Extended translational symmetry in the theory of multisublattice magnets. Spin waves, phase transitions, and domain walls in antiferromagnets of type UX_n

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A new method is proposed for a theoretical analysis of multisublattice magnets. In certain cases the method makes it possible, by an exact mathematical procedure, to decrease effectively the number of magnetic sublattices and thereby simplify considerably the development of both a linear and a nonlinear theory of spin waves. The capabilities of the method are illustrated by a calculation of the magnon spectrum and of the antiferromagnetic-resonance frequencies in $MnCl_4(NH_3)_2(CH_2)_3$ in the cubic phase of uranium compounds of type UX_n. A theory of orientational phase transitions in UX_n is constructed with the aid of this method. The energy and structure of domain walls in the cubic phase of UX_n are calculated. A criterion for the degeneracy of the magnon spectrum on the whole surface of the Brillouin zone is formulated.

1. INTRODUCTION

Practically all magnetically ordered crystals have several atoms per unit magnetic cell, i.e., are of the multisublattice type. Examples are rare-earth orthoferrites with 8 magnetic sublattices, rare-earth iron garnets with 32, the popular antiferromagnet $CuCl_2 \cdot 2H_2O$ with 4 sublattices, and many other. The microscopic theory of a magnet having more than two magnetic atoms per unit cell is technically extremely complicated. Considerable difficulties are encountered even in the problem of finding the spectrum of the elementary excitations of such a magnet, let alone analysis of nonlinear processes. These circumstances make it urgent to develop symmetry methods that simplify the theoretical analysis of multisublattice systems.

The use of symmetry methods in the theory of magnetically ordered crystals was started in Refs. 1 and 2. The main idea of these papers was a change from sublattice magnetizations to their linear combinations that transform in accordance with irreducible representations of the symmetry group of the crystal's paramagnetic phase. Many results in the theory of static, high-frequency, optical, and other properties of magnets were subsequently obtained along these lines. Notwithstanding the appreciable progress achieved by following Refs. 1 and 2, however, the technical difficulties in the case of multisublattice magnets are still quite great.

Additional possibilities in the theory of magnetically ordered crystals are uncovered by color symmetry,[3,4] which admits of independent transformations of the space and spin variables and a symmetry of approximate theoretical models. Color symmetry is described by spin space (color) groups.[3–5]

We shall be interested in the present paper in color symmetry because it permits extension of the translational-symmetry concept. It was shown in Ref. 4 that operations of type $\hat{T}\hat{U}$ ($\hat{T}$—translation, $\hat{U}$—rotation in spin space) generate an Abelian subgroup of a spin space group, and this subgroup is isomorphous to the translation group. It was proposed in Ref. 3, and later in Ref. 4, to use this group to calculate the energy of stationary states in magnetic spirals and to classify their stationary state.

In the present paper we propose, on the basis of the $\hat{T}\hat{U} \to \hat{T}$ isomorphism, a method that permits to lower effectively in certain cases, by an exact mathematical procedure, the number of magnetic sublattices and thereby simplify appreciably the development of a linear as well as nonlinear theory of multisublattice magnets.

2. EXTENDED TRANSLATIONAL SYMMETRY IN SPIN-WAVE THEORY

We consider first a magnet in which multiplication of the unit cell takes place on going from the paramagnetic to the magnetically ordered phase. In other words, let the translation $\hat{T}$ be contained in the symmetry group of the paramagnetic phase and be absent from that of the magnetically-ordered phase. The initial Hamiltonian of the magnet is, of course, invariant to $\hat{T}$. In the development of a spin-wave theory, linear or nonlinear, the $\hat{T}$-invariance is lost. The reason is that spin-wave theory is a theory of small oscillations about the ground state. The symmetry of the ground state is that of a magnetically ordered phase, and consequently does not contain $\hat{T}$. In practice, in development of a spin-wave theory it is customary to change to local coordinate frames for the atomic spin moments, i.e., to frames whose Z axis coincides with the direction of the spin in the ground state. The Hamiltonian, as a function of local spin coordinates, loses in the general case the $\hat{T}$ invariance. This makes it necessary to introduce several magnetic sublattices and quasiparticles of different species even if the unit cell of the initial paramagnetic phase contains one magnetic atom. The ensuing technical complications are obvious.

In addition, one can imagine a situation in which the translational symmetry is raised on going to local spin coordinates: the Hamiltonian becomes invariant to a translation absent in the paraphase group. For example, as will be made obvious below, this is possible in models of a magnet in which the symmetry element is a screw axis or a slip plane. Allowance for the additional translational symmetry in such a case would noticeably simplify the development of a spin-wave theory.

It is thus logical to raise the question of how the translational symmetry of a spin Hamiltonian changes, under various conditions, on going to local coordinate frames in spin space. An answer to this question is the following lemma. If the spin Hamiltonian and its ground state are invariant to the symmetry operation $\hat{T}\hat{U}$, where $\hat{T}$ is translation and $\hat{U}$ is rotation in spin space, then the Hamiltonian as a function of

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the local spin coordinates is invariant to $\hat{T}$. To prove the lemma we consider a magnet with a Hamiltonian

$$H=\frac{1}{2}\sum_{nm}A_{n,m}^{\alpha\beta}S_n^{\alpha}S_m^{\beta} \tag{1}$$

that is invariant to $\hat{T}\hat{U}$. The subscripts n and m in (1) number the magnetic atoms, the superscripts $\alpha,\beta = x, y, z$ number the spin components, S_n^{α} are the spin-projection operators, and $A_{n,m}^{\alpha\beta}$ is the interaction-constants matrix. The action of the translation $\hat{T}$ is the atom rearrangement $\hat{T}(r_n) = r_{n+1}$. The action of $\hat{U}$ is rotation in spin space $(\hat{U}S_n)^{\alpha} = U_{\alpha\gamma}S_n^{\gamma}$. Invariance to the operation $\hat{T}\hat{U}$ imposes then on the coefficients of Hamiltonian (1) the condition

$$A_{n+1,m+1}^{\alpha\beta}=A_{n,m}^{\gamma\mu}U_{\gamma\alpha}U_{\mu\beta}. \tag{2}$$

We introduce local coordinate frames for the atoms n and m. Then

$$S_n^{\alpha}=N_{\alpha\delta}\sigma_n^{\delta}, \quad S_m^{\beta}=M_{\beta\nu}\sigma_m^{\nu},$$

where σ_n^{δ} and σ_m^{ν} are the spin projections on the axes of the local systems, while $N_{\alpha\delta}$ and $M_{\beta\nu}$ are the matrices of the transformation from the crystallographic coordinate frames to the local ones. We introduce for the atoms $n+1$ and $m+1$ coordinate frames obtained by the rotation $\hat{U}$ from the local frames of the nth and mth atoms, respectively. Since the conditions of the lemma presuppose invariance of the ground state to $\hat{T}\hat{U}$, these frames will be local for the atoms $n+1$ and $m+1$:

$$S_{n+1}^{\alpha}=(U^{-1})_{\alpha i}N_{i\delta}\sigma_{n+1}^{\delta}, \quad S_{m+1}^{\beta}=(U^{-1})_{\beta j}M_{j\nu}\sigma_{m+1}^{\nu}.$$

The Hamiltonian of the interaction of the $(n+1)$st and $(m+1)$st atoms takes the form

$$\begin{aligned}H_{n+1,m+1}&=A_{n+1,m+1}^{\alpha\beta}S_{n+1}^{\alpha}S_{m+1}^{\beta}\\&=A_{n+1,m+1}^{\alpha\beta}(U^{-1})_{\alpha i}(U^{-1})_{\beta j}N_{i\delta}M_{j\nu}\sigma_{n+1}^{\delta}\sigma_{m+1}^{\nu}.\end{aligned}$$

Taking (2) into account, we have

$$\begin{aligned}H_{n+1,m+1}&=A_{n,m}^{\gamma\mu}U_{\gamma\alpha}U_{\mu\beta}(U^{-1})_{\alpha i}(U)_{\beta j}N_{i\delta}M_{j\nu}\sigma_{n+1}^{\delta}\sigma_{m+1}^{\nu}\\&=A_{n,m}^{\alpha\beta}N_{\alpha\delta}M_{\beta\nu}\sigma_{n+1}^{\delta}\sigma_{m+1}^{\nu}.\end{aligned} \tag{3}$$

The Hamiltonian of the interaction of the nth and mth atoms is

$$H_{n,m}=A_{n,m}^{\alpha\beta}N_{\alpha\delta}M_{\beta\nu}\sigma_n^{\delta}\sigma_m^{\nu}. \tag{4}$$

It is obvious from a comparison of (3) and (4) that in the local coordinate frames

$$\hat{T}H_{n,m}=H_{n+1,m+1}.$$

This proves the lemma.

Note that we used in the proof not arbitrary local frames, but such that are transformed into each other by the action of $\hat{T}\hat{U}$. Each $\hat{T}$ translation is accompanied here by a $\hat{U}$ rotation of the coordinate frames. The lemma is therefore valid only for just such a choice of the local spin coordinates.

Allowance for this remark makes obvious a method for a theoretical analysis of models of multisublattice magnets with properties that meet the conditions of the lemma. In this method, which can naturally be called the method of extended translational symmetry (ETS), $\hat{T}\hat{U}$-invariant local frames are chosen and the ensuing translational symmetry is successively used.

We emphasize once more that $\hat{T}\hat{U}$ invariance, and hence the possibility of using the ETS method, is obtained for definite model-dependent approximation.

In this connection, the exchange approximation[6,7] is worthy of a separate consideration. The exchange Hamiltonian is invariant to any rotation in spin space, so that in the exchange approximation the translational symmetry remains unchanged on going over to local spin coordinates.

In addition, from the standpoint of the ETS method, interest attaches to low-dimensionality models. The point is that in such models the conditions of the lemma are relaxed. For a one-dimensional chain it suffices to stipulate invariance of the Hamiltonian and of the ground state relative to a screw axis. Only if the positions of the magnetic atoms of the chain lie on a screw axis does its action reduce to $\hat{T}\hat{U}$. In the general case a screw axis is not $\hat{T}\hat{U}$, since it contains besides rotation in spin space also rotation of the atom coordinates. For one-dimensional models, however, only one coordinate is important, the one of the atom position along the chain. Variation of this coordinate by the action of a screw axis is translation. As applied to one-dimensional systems, the lemma can therefore be reformulated as follows.

If the Hamiltonian and the ground state of a one-dimensional magnetic chain are invariant relative to the screw axis, the Hamiltonian, as a function of the local spin coordinates, is invariant to the translation accompanying the screw axis.

For two-dimensional moments, the invariance to $\hat{T}\hat{U}$ can be replaced by invariance relative to a slip plane.

3. SPIN WAVES IN URANIUM COMPOUNDS

To illustrate the foregoing considerations we consider antiferromagnetic crystals of UO_2 and of the compound UX(X = N, P, As, Sb) that has been attracting interest of late.[3-11] We designate these crystals by UX_n. The space symmetry of UX_n is described by the group $O_h^5 - Fm3m$. The U atoms are located at inversion centers and form a face centered cubic (fcc) lattice. The corresponding Hamiltonian is

$$\begin{aligned}H=\frac{1}{2}\sum_n\Big\{&-\sum_{n'}I_{nn'}\mathbf{S}_n\mathbf{S}_{n'}+\sum_m J_{nm}\mathbf{S}_n\mathbf{S}_m-L_{nm}S_n^xS_m^x\\&+Q_{nm}(\mathbf{S}_n\mathbf{S}_m)^2+\sum_l J_{nl}\mathbf{S}_n\mathbf{S}_l-L_{nl}S_n^yS_l^y+Q_{nl}(\mathbf{S}_n\mathbf{S}_l)^2\\&+\sum_t J_{nt}\mathbf{S}_n\mathbf{S}_t\\&-L_{nt}S_n^zS_t^z+Q_{nt}(\mathbf{S}_n\mathbf{S}_t)^2\Big\}+\sum_n\{G((S_n^x)^4+(S_n^y)^4+(S_n^z)^4)\\&+D(S_n^x)^2(S_n^y)^2(S_n^z)^2\}.\end{aligned} \tag{5}$$

The unit cell of the paramagnetic phase contains one magnetic atom, the U atom, and the Hamiltonian (5) is invariant to the fcc lattice translations $\tau_x = (0, a/2, a/2)$, $\tau_y = (a/2, 0, a/2)$, $\tau_z = (a/2, a/2, 0)$, where a is the length of the edge of the face-centered cube. The subscript n numbers all the magnetic atoms. The subscripts m, l, and t number the neighbors of the n atom from the first coordination sphere in the planes YZ, ZX, and XY, respectively. The subscript n' numbers the neighbors of the atom also from the second coordination sphere.

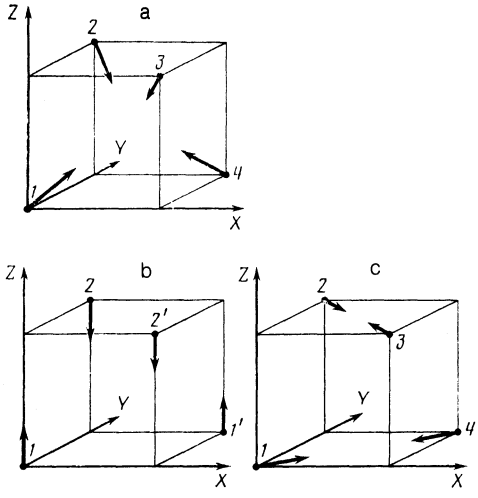

FIG. 1.

In the UX_n model it is of primary importance to take into account interactions of fourth order in the powers of the spins.[12] If only quadratic interactions are considered, the ground state of (5) becomes continuously degenerate with respect to a definite spin orientation in the magnetic cell. This degeneracy was lifted in the theoretical papers[12-14] devoted to UO_2 by using a model with biquadratic exchange. This is natural, since the exchange interaction is the strongest in the hierarchy of magnetic interactions. In heavy magnetic atoms such as U, however, the spin-orbit interaction is substantial, so that one-ion anisotropy is taken into account in (5) in addition to biquadratic exchange.

We consider below spin waves only in the cubic noncollinear phase of UX_n (Ref. 12). The cubic phase has the symmetry group $Pn3m'$. The corresponding magnetic unit cell contains for magnetic atoms or four crystallographic cells (see Fig. 1a). The numbers 1, 2, 3, and 4 in the figure designate the magnetic sublattices distinguished in the cubic phase.

Spin waves in the cubic phase of UO_2 were investigated by the traditional approach in Ref. 13. The Hamiltonian used there differed from (5) only in that the latter takes into account one-ion anisotropy but lacks the biquadratic exchange that connects triads and tetrads of atoms. In addition, the Hamiltonian of Ref. 13 is written in a form that explicitly postulates the presence of four magnetic sublattices. There is no need for this in principle, since the unit cell of the paraphase contains one magnetic atom, a circumstance taken into account in (5).

From the standpoint of the ETS method it is remarkable that the Hamiltonian (1) is invariant to the translations τ_x, τ_y, τ_z, and also to 180° rotations C_{2x}, C_{2y}, C_{2z} in spin space around the the axes X, Y, and Z.

The ground state of the cubic phase is invariant to the operations $\tau_x C_{2x}, \tau_y C_{2y}, \tau_z C_{2z}$. Therefore, according to the lemma, one can choose the coordinate frames for the spin moments in such a way that the Hamiltonian remains invariant to τ_x, τ_y, τ_z. In the unit magnetic cell the spin of the sublattice 1 is connected with the spins of sublattices 2, 3, and 4 by the operations $\tau_x C_{2x}, \tau_y C_{2y}, \tau_z C_{2z}$. We shall therefore use for the spins of sublattice 1 the initial crystallographic coordinate frame, and introduce for the spins of sublattices 2 , 3, and 4 coordinate frames obtained from the crystallographic by rotations around the axes X, Y, and Z, respectively.

We call these coordinate frames comoving. We note that comoving frames are not local in the sense that their Z axes do not coincide with the directions of the spins in the ground state. In the ground state, however, the projections on the axes of the comoving systems are the same for all spins. This is ensured by the fact that the systems are rotated by ground-state symmetry operations. If a transition is made for the spin of sublattice 1 from the crystallographic frame to the local one, the comoving frames for the remaining spins become automatically local.

Following the change to the comoving frame, the Hamiltonian (5) takes the form

$$H=\frac{1}{2}\sum_n\Big\{-\sum_{n'}I_{nn'}\boldsymbol{\sigma}_n\boldsymbol{\sigma}_{n'}+\sum_m J_{nm}(\sigma_n^x\sigma_m^x-\sigma_n^y\sigma_m^y-\sigma_n^z\sigma_m^z)$$
$$-L_{nm}\sigma_n^x\sigma_m^x+Q_{nm}(\sigma_n^x\sigma_m^x-\sigma_n^y\sigma_m^y-\sigma_n^z\sigma_m^z)^2+\sum_l J_{nl}(-\sigma_n^x\sigma_l^x$$
$$+\sigma_n^y\sigma_l^y-\sigma_n^z\sigma_l^z)-L_{nl}\sigma_n^y\sigma_l^y+Q_{nl}(-\sigma_n^x\sigma_l^x+\sigma_n^y\sigma_l^y-\sigma_n^z\sigma_l^z)^2$$
$$+\sum_t J_{nt}(-\sigma_n^x\sigma_t^x-\sigma_n^y\sigma_t^y+\sigma_n^z\sigma_t^z)$$
$$-L_{nt}\sigma_n^z\sigma_t^z+Q_{nt}(-\sigma_n^x\sigma_t^x-\sigma_n^y\sigma_t^y+\sigma_n^z\sigma_t^z)^2\Big\}$$
$$+\sum_n\{G((\sigma_n^x)^4+(\sigma_n^y)^4+(\sigma_n^z)^4)+D(\sigma_n^x)^2(\sigma_n^y)^2(\sigma_n^z)^2\}. \tag{6}$$

In the Hamiltonian (6), σ_n^α are the operators of the projections of the spin magnetic moments on the axes of the comoving systems:

$$\boldsymbol{\sigma}_n=(S_n^x, S_n^y, S_n^z),\quad \boldsymbol{\sigma}_m=(S_m^x, -S_m^y, -S_m^z),$$
$$\boldsymbol{\sigma}_l=(-S_l^x, S_l^y, -S_l^z),\quad \boldsymbol{\sigma}_t=(-S_t^x, -S_t^y, S_t^z). \tag{7}$$

Thus, the Hamiltonian (6) and its ground state are invariant to the translations τ_x, τ_y, τ_z,. Its analysis can be continued in the usual manner, by starting from one sublattice, and if necessary constructing a complete nonlinear spin-wave theory. In the present section we solve two problems: we obtain the spin-wave spectrum and the antiferromagnetic-resonance (AFMR) frequencies.

The ground state of (6) is given by

$$\sigma_x=\sigma_y=\sigma_z=S/3^{1/2},$$

where S is the spin. This is the known result that in the ground state the spins of UO_2 cubic phase are oriented along the body diagonals of a cube.

After a change to Bose operators and a Fourier transformation we obtain in the momentum representation the Hamiltonian component quadratic in the Bose operators:

$$H_2=\sum_k\{A_k b_k^+ b_k+{}^1/_2 B_k b_k b_{-k}+{}^1/_2 B_k^* b_k^+ b_{-k}^+\},$$
$$A_k=S[{}^8/_3 GS^2-{}^4/_9 DS^4+I_0(1-\gamma_k)+J_0+L_0+2Q_0S^2$$
$$-{}^1/_3(J_0+L_0-{}^{10}/_3 Q_0S^2)(\mu_{xk}+\mu_{yk}+\mu_{zk})],$$
$$B_k=\frac{S}{3}\left(J_0-\frac{L_0}{3}+\frac{2}{3}Q_0S^2\right)[(1+i\sqrt{3})\mu_{xk}$$
$$+(1-i\sqrt{3})\mu_{yk}-2\mu_{zk}], \tag{8}$$

where

$$I_0/I=6, \quad J_0/J=L_0/L=Q_0/Q=4,$$
$$\gamma_k=\frac{1}{3}(\cos ak_x+\cos ak_y+\cos ak_z), \quad \mu_{xk}=\cos\frac{ak_y}{2}\cos\frac{ak_z}{2}, \tag{9}$$
$$\mu_{yk}=\cos\frac{ak_z}{2}\cos\frac{ak_x}{2}, \quad \mu_{zk}=\cos\frac{ak_x}{2}\cos\frac{ak_y}{2}.$$

We obtain the spin-wave spectrum by the usual procedure of diagonalizing H_2:

$$\begin{aligned}\varepsilon_c^2(\mathbf{k})=&S^2[{}^8/_3GS^2-{}^4/_9DS^4+J_0+L_0+2Q_0+I_0(1-\gamma_k)\\&-{}^1/_3(J_0+L_0-{}^{10}/_3Q_0S^2)(\mu_{xk}+\mu_{yk}+\mu_{zk})]^2\\&-S^2\,{}^4/_9(J_0-{}^1/_2L_0+{}^2/_3Q_0S^2)^2(\mu_{xk}{}^2+\mu_{yk}{}^2+\mu_{zk}{}^2\\&-\mu_{yk}\mu_{zk}-\mu_{zk}\mu_{xk}-\mu_{xk}\mu_{yk}).\end{aligned} \tag{10}$$

We consider now resonance interaction between an external high-frequency field and spin waves. To obtain in closed form a theoretical scheme for this interaction in the framework of the ETS method, it is necessary to describe the magnetic field acting on the spin and the spin itself in one and the same coordinate frame. Therefore, to describe the high-frequency field we use the comoving coordinate frames (7). The field is no longer uniform with respect to these frames, since they have different orientations. We consider a specific situation in which a field of amplitude $\mathbf{h}_0$ is linearly polarized along the Z axis. The Z-projection of the field in the spin frames 1, 2, 3, and 4 (see Fig. 1a) are then h_0, $-h_0$, $-h_0$, h_0. This coordinate dependence can be represented in the form $\mathbf{h}(r,t)=\mathbf{h}(t)e^{i\mathbf{k}_0\mathbf{r}}$. In the case of Z-polarization we have $\mathbf{k}_0=\mathbf{k}_{0z}=(2\pi/a, 2\pi/a, 0)$. Such a field excites a magnon with momentum $\mathbf{k}_{0z}$. The resonance frequency is

$$\omega_z=\varepsilon_c(\mathbf{k}_{0z})=S\left[\frac{4}{3}\left(4GS^2-\frac{2DS^4}{3}+4J_0+L_0+\frac{8}{3}Q_0S^2\right)\times\left(4GS^2-\frac{2}{3}DS^4+L_0\right)\right]^{1/2}.$$

If the high-frequency magnetic field is polarized along the X and Y axes we have similarly $k_{0x}=(0, 2\pi/a, 2\pi/a)$, $k_{0y}=(2\pi/a, 0, 2\pi/a)$, and the resonance frequencies $\omega_x=\omega_y=\omega_z$, which is natural in view of the cubic symmetry.

Since we have reduced UX_n in the cubic magnetic phase to a system containing one U atom per magnetic cell, the spectrum (11) consists naturally of one mode. The quasimomentum varies then within a Brillouin zone corresponding to the translations τ_x, τ_y, τ_z. The usual approach yields a spectrum consisting of four modes, three acoustic and one exchange, but defined in a Brillouin zone whose volume is one-quarter as large. The different modes correspond here to different regions of the expanded band.

Comparing the spectrum (10) with the results of Ref. 13, it is easy to verify that the energies of magnons with momenta k_{0x}, k_{0y}, k_{0z} correspond to activation of the acoustic modes, and the energies of the magnons with momentum $k=0$ to activation of the exchange mode. Then

$$\omega_0=\varepsilon_c(k=0)={}^8/_3S^3(G-{}^1/_6DS^2+2Q_0).$$

A uniform external field does not excite magnons with momentum $\mathbf{k}=0$. This agrees with the conclusion of Ref. 13 that no UO_2 exchange mode is excited in AFMR.

4. ORIENTATIONAL PHASE TRANSITIONS IN UX_n

The possibilities of using the ETS method are not restricted to spin-wave theory. In fact, an additional translational symmetry appears in the ETS method prior to linearization or to introduction of some Bose representation. The ETS method can therefore be useful in the mean-field theory, in the Green's function method, etc.

We consider below, on the basis of the ETS method, the possible phases and orientational phase transitions in UX_n. An investigation of orientational phase transitions in UO_2 is of particular interest because different experimental papers contain contradictory data on its magnetic structure. In most studies, starting with Refs. 15 and 16, it is assumed that a cubic noncollinear magnetic phase is realized in UO_2. At the same time, earlier work[17–19] on inelastic neutron scattering in UO_2 point definitely to a tetragonal symmetry. This contradiction can be reconciled by assuming that a phase transition between the tetragonal and cubic phases can occur in UO_2 at a definite temperature.

The possible phases of UX_n were analyzed in Ref. 8, but without allowance for biquadratic exchange, which will be shown below to play a substantial role in the analysis of phase stability and phase transitions.

We note first that the comoving coordinate frames (7) used to calculate the magnon spectrum in the cubic phase permit a description of any magnetic phase whose symmetry group contains the operations $\tau_x C_{2x}$, $\tau_y C_{2y}$, $\tau_z C_{2z}$. The spin direction relative to the comoving frame uniquely parametrizes the phases of this aggregate. The direction corresponding to the cubic phase is then [11].

The thermodynamic-potential density at $T=0$ can be obtained in the quasiclassical approximation from the Hamiltonian (6), by identifying the nodes using the substitution $\boldsymbol{\sigma}_n\to\boldsymbol{\sigma}$:

$$\Phi=(G+2Q_0)[(\sigma^x)^4+(\sigma^y)^4+(\sigma^z)^4]+D(\sigma^x)^2(\sigma^y)^2(\sigma^z)^2. \tag{11}$$

It should be noted that the potential (11) receives no contribution from the terms quadratic in spin, and particularly from quadratic exchange, since the energy corresponding to them is independent of the spin direction. This important circumstance means that the barriers between the different phases are of low energy: they are determined only by one-ion anisotropy and biquadratic exchange. In addition, the constants of the potential (11), which correspond to interactions of different origin (one-ions anisotropy and biquadratic exchange), can vary differently with temperature, and this can lead in turn to phase transitions.

It is noteworthy that the thermodynamic potential (11) has the same structure as the potential of a cubic ferromagnet. One can therefore analyze (11) by using the results known for a cubic ferromagnet.[20]

Let us list the equilibrium phases of (11). The phase [111] is the already considered cubic noncollinear four-sublattice phase (see Fig. 1a). The condition for its stability is

$$G+2Q_0-DS^2/6\geqslant 0.$$

This result could have been obtained by considering the spectrum (10). Loss of stability of phase [111] corresponds

to softening of the magnons with momentum $\mathbf{k}=0$.

Phase [100] is a tetragonal collinear two-sublattice phase (see Fig. 1b). Its stability condition is

$$G+2Q_0\leqslant 0.$$

Also in equilibrium is the complanar four-sublattice phase [110] (Fig. 1c). Its stability condition is

$$0\leqslant G+2Q_0\leqslant DS^2/4.$$

A phase transition between [111] and [100] occurs under the condition

$$18(G+2Q_0)=DS^2.$$

It is evident that when the condition $D\leqslant 0$ is met the phase [110] is unstable. If, however, the stability condition for the [110] phase is met, the stability regions of [111] and [100] do not cross and a phase transition between them is impossible.

Since there is no experimental evidence of realization of a [110] phase in UO_2, it can be assumed that $D\leqslant 0$ in it.

A phase transition between [100] and [110] takes place under the condition

$$G+2Q_0=0, \quad D\geqslant 0.$$

A phase transition between [110] and [111] occurs if

$$9(G+2Q_0)=2DS^2, \quad D\geqslant 0.$$

All the phase transitions considered above are of first order.

Allowance for anisotropy and exchange of higher orders could yield a more detailed pictures of the magnetic phases and the transitions between them.

5. DOMAIN WALLS IN THE CUBIC PHASE OF UX_n

The ETS method can be useful also for the consideration of nonlinear inhomogeneous states in multisublattice magnets, particularly domain walls. We consider here domain walls of a special type in the cubic phase of UX_n, the existence of which was first pointed out by Dzyaloshinskiĭ.[21]

It was shown in Ref. 21, with UO_2 as the example, that orientational magnetic phases that can be transformed into one another by continuous rotation can exist in antiferromagnets with more than three sublattices. This pertains to rotations of the system of spin magnetic moments making up the magnetic unit cell, without disturbing the mutual orientation of the spins in the cell. The corresponding domain walls are not hydrodynamic, in the sense that they can be described in principle within the framework of a hydrodynamic approach[22] that operates with rotation of the spin "hedgehog" of the unit cell.

It was shown later[23] that, owing to the definite hierarchy of the interactions in UO_2, the domain-wall thickness can be macroscopic. This is possible because the mutual orientation of the spins in the cell is governed only by biquadratic exchange, which is much weaker than the quadratic exchange. The macroscopic character of the domain wall is in this case the result of competition between homogeneous biquadratic exchange, which tends to compress the wall, and inhomogeneous quadratic exchange, that tends to make the wall thicker.

Such domain walls can be obtained in principle as a solution of the system of Landau-Lifshitz equations for each of the four sublattices of the UO_2 cubic phase. The corresponding variational problem, however, would contain eight field variables and would be very complicated.

Yet the problem of domain walls in UX_n can be solved quite briefly by the ETS method. The point is that the energywise most important condition that determines the domain-wall structure in UX_n is that the energy of the homogeneous quadratic exchange be a maximum.[23] The Hamiltonian describing smooth variations of the spin magnetic moments in space can be obtained by expanding (6) in terms of small gradients:

$$H=\int\left\{(G+2Q_0)\left[(\sigma^x)^4+(\sigma^y)^4+(\sigma^z)^4\right]+\frac{J_0a^2}{8}\left[\left(\frac{\partial\sigma^x}{\partial x}\right)^2+\left(\frac{\partial\sigma^y}{\partial y}\right)^2+\left(\frac{\partial\sigma^z}{\partial z}\right)^2\right]\right\}dV. \tag{12}$$

The Hamiltonian (12) does not contain the homogeneous energy of either the cubic inter-ion anisotropy nor, more importantly, biquadratic exchange. In the walls described by (12), the principal energy condition is met automatically. This is precisely why (12) can be used to describe the domain walls in UO_2 in terms of one vector field $\boldsymbol{\sigma}(\mathbf{r})$.

Sixth-order anisotropy has been left out of the Hamiltonian (12) because it does not play a principal role in the analysis of domain walls far from the phase transitions. Also left out are the inhomogeneous terms corresponding to quadratic anisotropy and biquadratic exchange, since they are small compared with the inhomogeneous terms taken into account in (12).

We point out that the inhomogeneous part of the Hamiltonian (12) has a rather unusual, anisotropic form, although it is derived from a pure exchange Hamiltonian.

Eight different orientational domains are possible in the cubic phase. They are characterized by the following directions of $\boldsymbol{\sigma}$:

$$(1, 1, 1),\ (1, -1, -1),\ (-1, 1, -1),\ (-1, -1, 1), \tag{13}$$

$$(-1, -1, -1),\ (-1, 1, 1),\ (1, -1, 1),\ (1, 1, -1). \tag{14}$$

The domains belonging to any one set, (13) or (14), can be obtained from one another by rotating, without disturbing its internal arrangement, the "hedgehog" of the spin magnetic moments that form the unit cell. The corresponding domain walls can be obtained by the usual hydrodynamic approach. We do not consider such walls here.

Domains belonging to different sets (13) and (14) cannot be obtained from one another by rotations of the "hedgehog." On going from any domain from (13) to any domain from (14), the mutual orientation of the spins in the cell must change.[21] It is the corresponding domain walls which we treat in the present article.

Consider, by way of argument, the wall between the domains (1, 1, 1) and (1, 1, −1). We introduce for the components of the vector $\boldsymbol{\sigma}$ the notation

$$\sigma^x=S\sin\theta\cos\varphi, \quad \sigma^y=S\sin\theta\sin\varphi, \quad \sigma^z=S\cos\theta.$$

In the domain (1, 1, 1) we have

$$\theta=\theta_1=\arccos(1/\sqrt{3}), \quad \varphi=\varphi_0=\pi/4, \tag{15}$$

in the domain (1, 1, −1)

$$\theta=\theta_2=-\arccos(1/\sqrt{3}), \quad \varphi=\varphi_0=\pi/4. \tag{16}$$

We seek the solution in the form of a plane wall. We introduce for this purpose a spatial coordinate defined by the relation

$$x=n_x\xi, \quad y=n_y\xi, \quad z=n_z\xi,$$

where n_x, n_y, and n_z are the direction cosines of the normal to the wall plane, and assume that θ and φ are functions of ξ only. The Hamiltonian (12) takes then the form

$$H=\int dV\Big\{(G+2Q_0)S^4[\cos^4\theta+\sin^4\theta(\cos^4\varphi+\sin^4\varphi)] + \frac{J_0a^2S^2}{8}\Big(\frac{1}{n_x^2}\Big(\cos\varphi\cos\theta\frac{\partial\theta}{\partial x}-\sin\varphi\sin\theta\frac{\partial\varphi}{\partial x}\Big)^2 + \frac{1}{n_y^2}\Big(\sin\varphi\cos\theta\frac{\partial\theta}{\partial y}+\cos\varphi\sin\theta\frac{\partial\varphi}{\partial y}\Big)^2 + \frac{1}{n_z^2}\Big(\sin\theta\frac{\partial\theta}{\partial z}\Big)^2\Big)\Big\}. \tag{17}$$

Solution of the variational problem for the Hamiltonian (17) with boundary conditions (15) and (16) leads to

$$\cos\theta(z)=\frac{1}{\sqrt{3}}\operatorname{th}\frac{z-z_0}{\lambda}, \quad \varphi=\frac{\pi}{4}, \quad n_z=1,$$

where

$$\lambda=\frac{a}{S}\Big(\frac{J_0}{G+2Q_0}\Big)^{1/2}$$

is the domain-wall thickness. An important feature is that the boundary conditions (15) and (16) determine uniquely the orientation of the wall in space. The normal to the wall between the domains (1, 1, 1) and (2, 1, −1) is directed along the Z axis.

The domain-wall surface tension is

$$\Delta=^8/_9 aS^3[J_0(G+2Q_0)]^{1/2}.$$

6. SPIN WAVES IN $MnCl_4(NH_3)_2(CH_2)_3$

As an example of a magnet whose translational symmetry increases on going to local spin coordinates we consider the quasi-twodimensional four-sublattice antiferromagnet $MnCl_4(NH_3)_2(CH_2)_3$. This crystal has been recently attracting intent attention in view of the magnetic-resonance modes and unusual pattern of phase transitions observed in it.[24,25] The paramagnetic-phase symmetry of this antiferromagnet is described by the group D_{2n}^{16}. The magnetic atoms are located at inversion centers. The $MnCl_4(NH_3)_2(CH_2)_3$ crystal constitutes an assembly of monoclinic antiferromagnetic layers weakly coupled to one another by antiferromagnetic exchange. The presence of a second-order screw axis unites the monoclinic layers into a rhombic crystal. We consider an $MnCl_4(NH_3)_2(CH_2)_3$ model that takes into account only exchange interactions and one-ion anisotropy, $H=H_1+H_2+H_{1,2}$, where

$$H_1=\sum_{n_1,n_3}J_{n_1,n_3}\mathbf{S}_{n_1}\mathbf{S}_{n_3}-\frac{1}{2}\beta^z[(S_{n_1}^z)^2+(S_{n_3}^z)^2] - \frac{1}{2}\beta^y[(S_{n_1}^y)^2+(S_{n_3}^y)^2] - \frac{\alpha}{4}(S_{n_1}^yS_{n_1}^z+S_{n_1}^zS_{n_1}^y+S_{n_3}^yS_{n_3}^z+S_{n_3}^zS_{n_3}^y),$$

$$H_2=\sum_{n_2,n_4}J_{n_2n_4}\mathbf{S}_{n_2}\mathbf{S}_{n_4}-\frac{1}{2}\beta^z[(S_{n_2}^z)^2+(S_{n_4}^z)^2] - \frac{1}{2}\beta^y[(S_{n_2}^y)^2+(S_{n_4}^y)^2] + \frac{\alpha}{4}(S_{n_2}^yS_{n_2}^z+S_{n_2}^zS_{n_2}^y+S_{n_4}^yS_{n_4}^z+S_{n_4}^zS_{n_4}^y),$$

$$H_{1,2}=\sum_{n_1,n_2}I_{n_1,n_2}\mathbf{S}_{n_1}\mathbf{S}_{n_2}+\sum_{n_3,n_4}I_{n_3,n_4}\mathbf{S}_{n_3}\mathbf{S}_{n_4}. \tag{18}$$

The subscripts n_1, n_2, n_3, n_4 in (18) number magnetic atoms belonging to four different sublattices. The Hamiltonians H_1 and H_2 describe interaction in the layers, with J the intralayer antiferromagnetic-exchange constant, and β^z, β^y, and α the one-ion anisotropy constants. The Hamiltonian $H_{1,2}$ describes interlayer interaction and I is the interlayer antiferromagnetic-exchange constant. The crystal $MnCl_4(NH_3)_2(CH_2)_3$ is quasi-two-dimensional because $J\gg I\sim\alpha,\beta^z,\beta^y$. Equation (21), of course, does not take into account all the invariants of the D_{2n}^{16} group. In particular, no account is taken in (18) of the Dzyaloshinskiĭ interaction, which forms the weak ferromagnetic moment. As shown in Ref. 24, however, the Hamiltonian (18) permits an adequate description of many observed properties of this crystal.

The magnetic unit cell of $MnCl_4(NH_3)_2(CH_2)_3$ is shown in Fig.2, where the numbers *1*, *2*, *3*, and *4* mark the magnetic sublattices. The translational symmetry of the magnetic phase is specified by the basis $t_x=(a, 0, 0)$, $t_y=(0, b, 0)$, $t_z=(0, 0, c)$, where a, b, and c are the lengths of the edges of the rhombic base-centered cell.

At the same time, the Hamiltonian (18) and its ground state are invariant to the operations τ_xC_{2x}, τ_yC_{2y}, where $\tau_x=(a/2, b/2, 0)$, $\tau_z=(0, 0, c/2)$, while C_{2x} and C_{2y} are 180° rotations in spin space around the X and Y axes, respectively. According to the lemma, this means that one can choose for the spins local coordinate frames such that the Hamiltonian (18) becomes invariant to the translations and τ_x and τ_y.

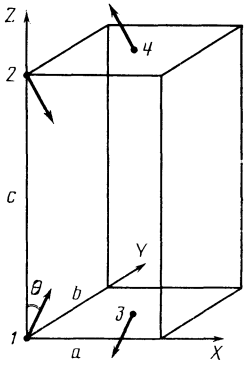

FIG. 2.

It should be noted here that the translation τ_z is a symmetry operation for neither the ground state nor the Hamiltonian. Thus, on going to the local spin coordinates the translational symmetry is increased compared with the paraphase.

We introduce appropriate coordinate frames for the sublattice spins. We use for sublattice *1* the crystallographic frame, from which we obtain for the spins of sublattices *2*, *3*, and *4* comoving frames by means of the rotations C_{2y}, C_{2x} and C_{2z}. This choice of the comoving frames is determined by the fact that the spin of sublattice *1* is connected with the spins of sublattices *2*, *3*, and *4* by the respective operations $\tau_z C_{2y}$, $\tau_x C_{2x}$ and $\tau_x \tau_z C_{2x} C_{2y}$. Then

$$H=\frac{1}{2}\sum_n\Big\{\sum_m J_{nm}(\sigma_n^x\sigma_m^x-\sigma_n^y\sigma_m^y-\sigma_n^z\sigma_m^z)+\sum_l I_{nl}(-\sigma_n^x\sigma_l^x+\sigma_n^y\sigma_l^y-\sigma_n^z\sigma_l^z)-\beta^z(\sigma_n^z)^2-\beta^y(\sigma_n^y)^2-\frac{\alpha}{2}(\sigma_n^y\sigma_n^z+\sigma_n^z\sigma_n^y)\Big\}. \quad (19)$$

In (19), σ_n^α are the operators of spin projection on the axes of the comoving systems:

$$\sigma_n=(S_{1n}^x,\ S_{1n}^y,\ S_{1n}^z),\quad \sigma_l=(-S_{2l}^x,\ S_{2l}^y,\ -S_{2l}^z),$$
$$\sigma_m=(S_{3m}^x,\ -S_{3m}^y,\ -S_{3m}^z),\quad \sigma_t=(-S_{4t}^x,\ -S_{4t}^y,\ S_{4t}^z).$$

The subscript n numbers all the magnetic atoms, m the nearest neighbors of the atom of n in the layer, and l the nearest neighbors of n from other layers.

The four-sublattice antiferromagnet has thus been reduced to a one-sublattice system.

Minimizing (19), we find that the angle θ between the ground-state spin direction and the Z axis of the crystallographic frame is given by

$$\operatorname{tg} 2\theta=\alpha/(2I_0+\beta^z-\beta^y). \quad (20)$$

The spin-wave spectrum corresponding to the Hamiltonian is

$$\varepsilon^2(\mathbf{k})=S^2[J_0(1+\gamma_k)+I_0(\cos\theta-\mu_k)+\delta_1]\times[J_0(1-\gamma_k)+I_0\cos\theta(1+\mu_k)+\delta_2],$$

where

$$J_0=4J,\quad I_0=2I,\quad \gamma_k=\cos\frac{k_xa}{2}\cos\frac{k_ya}{2},\quad \mu_k=\frac{\cos k_zc}{2},$$
$$\delta_1=\beta^z\cos^2\theta+\beta^y\sin^2\theta+{}^1/_2\alpha\sin 2\theta,$$
$$\delta_2=(\beta^z-\beta^y)\cos 2\theta+\alpha\sin 2\theta.$$

Interaction with a high-frequency field polarized along the axes X, Y, and Z excites magnons with momenta

$$\mathbf{k}_{0x}=(0,\ 0,\ 2\pi/c),\quad \mathbf{k}_{0y}=(0,\ 2\pi/b,\ 0),\quad \mathbf{k}_{0z}=(2\pi/a,\ 0,\ 2\pi/c).$$

The corresponding resonance frequencies are

$$\omega_x=\varepsilon(\mathbf{k}_{0x})=S\{[2J_0+I_0(\cos 2\theta+1)+\delta_1]\,\delta_2\}^{1/2},$$
$$\omega_y=\varepsilon(\mathbf{k}_{0y})=S\{(2J_0+2I_0\cos 2\theta+\delta_2)\,[\delta_1-I_1(1-\cos 2\theta)]\}^{1/2},$$
$$\omega_z=\varepsilon(\mathbf{k}_{0z})=S\{(2J_0+\delta_2)\,[I_0(\cos 2\theta+1)+\delta_1]\}^{1/2}.$$

Interest attaches also to the point $\mathbf{k}=0$, corresponding to the frequency

$$\omega_0=\varepsilon(\mathbf{k}=0)=\{[2J_0-I_0(1-\cos 2\theta)+\delta_1](2I_0\cos 2\theta+\delta_2)\}^{1/2}.$$

The frequency ω_z corresponds to the exchange mode, and ω_x and ω_y to the acoustic modes of the magnetic resonance. These frequencies correspond to the homogeneous oscillation modes obtained in Ref. 24.

7. CONCLUSION

In all the examples considered, the magnetically ordered crystals were described by a model Hamiltonian of rather general form. Besides the exchange interaction, account was taken of various relativistic interactions. We have thereby succeeded in reducing four-sublattice antiferromagnets to single-sublattice systems. In our opinion, this demonstrates clearly the extensive capabilities of the ETS method.

Mention must be made of several studies devoted to two-sublattice antiferromagnets[26,27] and spiral structures,[28,29] where use was made of the conservation of the translational symmetry of the Hamiltonian after changing to local coordinate frames. Analysis shows that in all these papers the Hamiltonian and the ground state satisfy the conditions of the lemma formulated by us.

We note in conclusion that the lemma has one more aspect. It is well known that the energy spectrum of spin waves can become degenerate in symmetry points of the Brillouin zone. It is obvious from our analysis that when the conditions of the lemma are met degeneracy appears on entire surfaces of the Brillouin zone. We have in mind here, of course, the usual Brillouin zone, and not the one obtained in the ETS method. This circumstance is a direct consequence of the isomorphism $\hat{T}\hat{U}\to\hat{T}$ (Ref. 4).

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